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Phosphorus loadings to the world's largest lakes: sources and trends

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Key Points

1. Phosphorus loadings to lakes are a major cause of lake eutrophication, yet there are few estimates of the extent of these loadings, including for the world's largest lakes.
2. TP loadings to large lakes in developing countries are on the average much larger than to lakes in developed countries.
3. The most important source of TP loadings is inorganic fertilizer; the second most important are "background" loadings from atmospheric deposition and catchment weathering of P.

4. Phosphorus loadings to large lakes increased in 50 out of 100 lakes between 1990-1994 and 2005-2010.

Abstract

Eutrophication is a major water quality issue in lakes worldwide and is principally caused by the loadings of phosphorus from catchment areas. It follows that to develop strategies to mitigate eutrophication we must have a good understanding of the amount, sources, and trends of phosphorus pollution. This paper provides the first consistent and harmonious estimates of current phosphorus loadings to the world's largest 100 lakes, along with the sources of these loadings and their trends. These estimates provide a perspective on the extent of lake eutrophication worldwide, as well as potential input to the evaluation and management of eutrophication in these lakes. We take a modelling approach and apply the WorldQual model for these estimates. The advantage of this approach is that it allows us to fill in large gaps in observational data. About 66 of the 100 lakes are located in developing countries and these have a much larger average loading than the lakes in developed countries (11.1 vs. 0.7 kg km^{-2} yr^{-1}). The main source of phosphorus to the examined lakes is inorganic fertilizer (47% of total). Between 2005-2010 and 1990-1994, phosphorus loadings increased to 50 out of 100 lakes; this included 30 out of 66 lakes in developing countries. Phosphorus loadings decreased or remained the same in 14 out of 44 lakes in developed countries. The average change in phosphorus loading was an increase of 7%, but this varied substantially between lakes. Trends in loadings in both directions were caused primarily by changing use of inorganic fertilizer.

Index Terms and Keywords

1871, 1806, 1845, 0496, 0458

phosphorus loadings, water quality, lake eutrophication, lakes, WorldQual model, WaterGap Modelling Framework

1 Introduction

Eutrophication is considered as one of the most important forms of lake pollution because of the extent of its occurrence and the intensity of its impact on lake water quality (*Millennium Ecosystem Assessment*, 2005). Eutrophication is an oversupply of nutrients to an aquatic system, usually causing undesirable changes in aquatic ecosystems such as toxic algal blooms, decrease in water transparency, oxygen depletion or anoxia due to decomposition of organic matter, changes in species composition, increased incidence of fish kills, reduced species diversity, and a reduction in harvestable fish (*Correll*, 1998; *Smith & Schindler*, 2009). Eutrophication threatens freshwater ecosystem services important to human well-being such as the provision of drinking water, the harvesting of fish, and the recreational use of lakes (*Aylward et al.*, 2005). For instance, in Lake Taihu, China, eutrophication in the form of massive algal blooms eutrophication has endangered the drinking water supply of millions of people in Shanghai and other cities (*Qin et al.*, 2007). Another well-known example is Lake Erie (Canada and USA) where the discharge of agricultural, industrial and domestic wastewater stimulated algal blooms, and fish kills until the mid-1980s (*Allinger & Reavie*, 2013). It takes up to decades for lakes to recover from eutrophication (*Jeppesen et al.*, 2005). Solving eutrophication problems was given new attention in 2015, when UN Member States agreed “By 2020, [to] protect and restore water-related ecosystems, including … lakes” as part of Target 6.6 of the UN Sustainable Development Goals (*United Nations*, 2015).

To assess the risk of eutrophication in large lakes, it is important to understand the loadings of phosphorus (P) to these lakes from their catchment. There are two reasons for this. Firstly, P is commonly the main limiting nutrient for phytoplankton growth in freshwater systems (*Sterner*, 2008; *Dodson*, 2005; *Brönmark & Hansson*, 2005); hence a certain level of P in lakes above normally pristine levels is a common trigger of eutrophication (*Schindler*, 1977; *Correll*, 1998;

Yan et al., 2016). Secondly, P loadings into a lake also affect the balance of other nutrients, such as nitrogen, that play a role in phytoplankton production (e.g. *Finlay et al.*, 2013). Because of the importance of P, many investigators have measured P loadings to lakes. A variety of methods have been used including pollution source assessments (e.g. *Scheren et al.*, 2000) and measurements of inflows (e.g. *Zimmer & Bendoricchio*, 2001). Estimates of P loadings to various lakes around the world have been compiled in the World Lake Database (*ILEC*, 2015) and for Europe in *European Environment Agency* (2005). Specifically for large lakes, estimates of P loadings have been published for Lake Victoria (*Scheren et al.*, 2000), Lake Michigan (*Johengen et al.*, 1994), Lake Baikal (*Callender & Granina*, 1997), Lake Malawi (*Pasche et al.*, 2012), Lake Erie (*Dolan & McGunagle*, 2005), Lake Winnipeg (*LWSB*, 2006), Lake Ladoga (*Holopainen & Letanskaya*, 1999), and Lake Onega (*Bilaletdin et al.*, 2011). Some of these studies, e.g. *Scheren et al.* (2000), provide information on P sources as well. Hence, there are already estimates of current P loadings to several large lakes.

However, in this paper we present for the first time a consistent estimate of total phosphorus (TP) loadings to the world 100 largest lakes, by catchment area. As a tool we use the WorldQual model from the global modeling framework WaterGAP3 (*Verzano*, 2009; *Flörke et al.*, 2013; *aus der Beek et al.*, 2010; *Voß et al.*, 2012; *Alcamo et al.,* 2003). These new top-down, model-based calculations provide added-value to current P loading estimates in the following ways: First, they cover many more large lakes and in a more consistent way than previous research; Second, using the same model to estimate P loadings to several lakes enable consistent comparisons between lakes because a uniform method is used for calculations; It is difficult to compare literature values for different lakes because they have used different methods (noted above) and different data inputs. Third, using a deterministic mass-balance model driven by various sources of P allows the linkage between sources and resulting loadings. This provides important information to policymakers and lake managers because it makes it possible to identify which sources must be reduced to substantially reduce loadings. Fourth, the model can be used to estimate changes in the sources of P within a lake basin, and hence be used to generate a time series of historic loadings. Fifth, the model can be used to make projections of changes in P loadings and hence provide input to assessments of changes in the intensity of problems as a function of socio-economic drivers and climate change. Likewise, it can be used to compute scenarios of progress towards achieving targets, such as those under the Sustainable Development Goals.

In this paper, we aim to answer three main questions:

(A) What are the magnitudes of P loadings to large lakes worldwide, and how do they vary between lakes?
(B) What are the main sources of P loading to large lakes and how do patterns vary worldwide?
(C) What are the twenty-year trends of P loadings to large lakes?

2 The investigated lakes

In this study we focus on the world's major lakes, which we define as the 100 largest lakes and reservoirs in terms of lake surface area, as identified in the Global Lake and Wetland Database (*Lehner & Döll*, 2004). We exclude the Caspian Sea (technical limitation for its size) and the Aral Sea and Lake Chad (rapidly changing morphology). Hereafter, for the sake of simplicity we use the term 'lake' for reservoirs.

Although most lakes are small, the few with large volumes and/or surface areas are especially significant because of their unique ecosystems and large fisheries. In some cases they are also a major supplier of water for irrigation, industry and the domestic sectors. The 100 largest lakes cover a wide range of environmental conditions and levels of water quality. Some lakes in the north are covered with ice for more than half the year, while the tropical lakes of South America never freeze. Anthropogenic loads of nutrients are higher in intensively farmed, industrial, and semi-rural or urban areas (e.g. Lake Taihu; *Wang et al.*, 2014), and much lower in remote catchments (e.g. Great Slave Lake, Canada).

The large lakes investigated here consists of 11 lakes in Europe (EU), 17 in Africa (AF), 28 in Asia (AS), 1 in Oceania (AU), 33 in North and Central America (NA), and 10 in South America (SA). The lake's mapping to each continent is defined by the land mask used in the WaterGAP3 modelling framework (Figure 1). For a detailed list of the studied lakes, see Supporting Information.

3 Methods

3.1 Modelling framework WaterGAP3

The main tool used in this study is the WorldQual model of the WaterGAP3 modelling framework (Water – Global Assessment and Prognosis). WaterGAP3 is a grid-based, integrative assessment tool operating on 5 arc minute global grid (about 9 by 9 km at the equator) (*Alcamo* et al., 2003; *Verzano*, 2009; *Flörke et al.*, 2013; *aus der Beek et al.*, 2010; *Voß et al.*, 2012) which has been applied in numerous studies (e.g. *Ward et al.*, 2014; *Williams et al.*, 2012; *Reder et al.*, 2013). The modelling framework (Figure 2a) includes three modules: (i) The distributed global hydrological model (*Alcamo et al.*, 2003; *Döll et al.,* 2003; *Eisner*, 2016) simulates hydrological storage compartments with a daily temporal resolution. The hydrological model is driven by a WFDEI meteorological data set (*Weedon et al.*, 2014) and calibrated and validated against measured river discharge from 2446 GRDC stations (*Eisner*, 2016). (ii) The water use module includes five sectoral water use models (*Flörke et al.*, 2013; *aus der Beek et al.*, 2010). (iii) The water quality model WorldQual (*Voß et al.*, 2012) simulates monthly loadings and in-stream concentrations from point sources and diffuse sources (Figure 2b) of total phosphorus (TP), fecal coliform bacteria (FC), total dissolved solids (TDS), and biochemical oxygen demand (BOD) (*Punzet et al.*, 2012; *Voß et al.*, 2012; *Williams et al.* 2012; *Reder et al.*, 2015).

Using several large global data sets (Table 1), the WorldQual model calculates TP loadings into lakes in two major steps. First, the TP loadings in each grid cell j in the lake catchment are calculated and summed. This includes loadings from domestic sewered wastewater ($Ld_{ds\mathrm{TP},j}$), domestic non-sewered wastewater ($Ld_{dns\mathrm{TP},j}$), manufacturing wastewater ($Ld_{mf\mathrm{TP},j}$), urban surface runoff ($Ld_{usr\mathrm{TP},j}$), inorganic fertilizer ($Ld_{if\mathrm{TP},j}$), and livestock wastes ($Ld_{org\mathrm{TP},j}$). The methods presented in sections 3.1.1– 3.1.7 are used for these calculations. The sum of all cell loadings in a lake catchment is:

$$L_{sumTP}[\mathrm{t\ month^{-1}}] = \sum_{j=1}^{n} Ld_{ds\mathrm{TP},j} + Ld_{dns\mathrm{TP},j} + Ld_{mf\mathrm{TP},j} + Ld_{usr\mathrm{TP},j} + Ld_{if\mathrm{TP},j} + Ld_{org\mathrm{TP},j} \quad (1)$$

L_{sumTP} represents the sum of all anthropogenic TP generated in the catchment. In the second step, the amount of these loadings retained in rivers, smaller lakes, and wetlands in the catchment is calculated (section 3.1.8). The remainder of the loading is assumed to drain into the main lake of the catchment.

As compared to other large-scale models, WorldQual and the WaterGAP3 framework have global coverage and computes nutrient source terms on a global 5 arc minute grid; it therefore can depict the spatial variability of pollution sources. Other models compute nutrient export at the mouth of rivers [e.g. the lumped models NEWS2 (*Mayorga et al.*, 2010), NANI/NAPI (*Hong et al.*, 2012)], or only for selected large scale basins [e.g. the semi-distributed and distributed models RiNUX (*Loos et al.*, 2009), HBV-NP (*Andersson et al.*, 2005)]. In this paper we use WorldQual to compute the P loading on a grid cell basis and then sum up to a lake basin total, whereas we compute P retention as a basin-average. The methodology for calculating TP loadings for various sectors is described in the following sections.

3.1.1 Domestic sewered wastewater

The TP loading from domestic sewered wastewater is the phosphorus in domestic wastewater that is collected in sewers but not removed by wastewater treatment. This loading is calculated as:

$$Ld_{ds\mathrm{TP}}[\mathrm{t\ TP\ month}^{-1}] = Pc/12 \times N/Pc \times Ex \times P/N \times Pop \times Cr \times (1 - Tr) \qquad (2)$$

where *Pc* [t cap^{-1} yr^{-1}] is the country-specific protein consumption per capita and year. Data for *Pc* were obtained from the *Food and Agriculture Organization* (*FAO*, 2014). For countries with no *Pc* information continental averages were taken. N/*Pc* is the average fraction of Nitrogen (N) in consumed protein (16%). *Ex* is the ratio of excreted/consumed protein (36.5% on average, *Van Drecht et al.*, 2009), P/N is the ratio of P and N in human feces (17% on average, *Van Drecht et al.*, 2009). *Pop* is the population in each grid cell (HYDE data base, *Klein Goldewijk et al.*, 2010). *Cr* [0-1] is the fraction of people living in the grid cell that are connected to the sewage system. *Tr* is the country-average treatment level for TP removal in sewage treatment plants. This factor accounts for removal in primary, secondary, and tertiary treatment and also accounts for the deficiencies in achieving the design removal rate. Data for *Cr* and Tr are from *Reeder* (2017).

3.1.2 Domestic non-sewered waste water (scattered settlements)

"Domestic non-sewered waste water from scattered settlements" consists of waste or wastewater that is collected at onsite disposal facilities (for example in septic tanks or hanging latrines), and then after treatment, or not, is finally introduced to surface waters as point sources. It also consists of wastes from open defecation and pit latrines that are washed off land surfaces by precipitation and enter lakes as a diffuse source. In WorldQual TP loadings from scattered settlements ($Ld_{dns\mathrm{TP}}$) are estimated similarly to equation (2) but also takes into account the fraction of urban and rural population that is not connected to the sewage system. To distinguish between diffuse and point sources from scattered settlements, data on sanitation practices are derived from the Joint Monitoring Programme (JMP) for water supply and sanitation (WHO/UNICEF, 2013), national databases, and the literature.

3.1.3 Wastewater from the manufacturing sector

Wastewater from manufacturing facilities is assumed to be collected in a sewage system and transported to wastewater treatment facilities where part of its phosphorus content is removed. The remainder is discharged to the local surface water system. TP loadings originating from manufacturing wastewater are calculated from:

$$Ld_{mf\mathrm{TP}}[\mathrm{t\ TP\ month^{-1}}] = C_{\mathrm{TP},mf} \times 10^{-9} \times Rfl_{mf}/12 \times (1 - Tr) \tag{3}$$

where $C_{\mathrm{TP},mf}$ [mg TP L^{-1}] is the average TP concentration in manufacturing wastewater. WorldQual uses a value of 3 mg TP L^{-1} as a average value over literature values for different manufacturing sectors. Rfl_{mf} [L yr^{-1}] is the return flow from the manufacturing industry in each grid cell and month as calculated by the Water Use model of WaterGAP3 (*Flörke et al.*, 2013; *aus der Beek et al.*, 2010). Similar to the domestic sector, manufacturing loads are reduced by the country-average treatment removal rate, *Tr* [0-1].

3.1.4 Urban surface runoff

Some phosphorus accumulates on urban surfaces and is washed off to surface waters by precipitation to sewage canals or other drainage routes. Sewage treatment plants are assumed to remove P from urban surface runoff at the same rate as they remove P from domestic sewered wastewater (see section 3.1.1). Monthly TP loadings in urban surface runoff are calculated by multiplying an event mean concentration (EMC_{TP} [mg L^{-1}]) times the monthly urban surface runoff R_{us} [mm month^{-1}]:

$$Ld_{usr\mathrm{TP}}[\mathrm{t\ TP\ month}^{-1}] = R_{us} \times EMC_{\mathrm{TP}} \times A_{cell} \times 10^{-3} \times F_{built} \times (1 - Tr) \tag{4}$$

A_{cell} [km²] and F_{built} [0-1] are the cell area and built up fraction, respectively. In this study, R_{us} is from the WaterGAP3 calculations presented in (*Schellekens et al.*, 2017). For EMC_{TP} we use a representative value of 0.2 mg L^{-1} based on *Göbel et al.* (2007).

3.1.5 Inorganic fertilizer in the agricultural sector

Our calculations also take into account the fraction of P contained in inorganic fertilizer which is not taken up by plants but is washed instead into the surface water system (in this paper we use "inorganic fertilizer" to mean manufactured fertilizer). We estimate this loading by multiplying the applied inorganic fertilizer $F_{\mathrm{inorg,TP}}$ [t TP yr^{-1}] times the terms in brackets that account for the loss of P by leaching and erosion.

$$Ld_{ifTP}[\mathrm{t\ TP\ month}^{-1}] = F_{\mathrm{inorg,TP}} \times \left(\frac{L_{max}}{1 + (R_{act}/a)^{-b}} + Sl \times c \times \frac{R_{act}}{R_{mean}}\right) \times 1/12 \tag{5}$$

L_{max} [-] is the maximal leachable fraction of TP in applied inorganic fertilizer TP (dissolved fraction). R_{act}/R_{mean} represents a correction factor that includes the annual variation of surface runoff (R_{act} [mm yr^{-1}] is the surface runoff in a year, R_{mean} [mm yr^{-1}] is the mean annual surface runoff 1980-2000). R_{act} and R_{mean} are calculated by the WaterGAP 3 hydrology model and correspond to the WaterGAP3 estimates in a global water resources reanalysis (*Schellekens et al.*, 2017). a [mm] and b [-] define the shape of the response of dissolved TP loadings on runoff changes. Their values as well as the value for L_{max} were taken from *Harrison, et al.* (2005)

(a=850mm, b=2, L_{max}=0.04). Sl is the gridded soil loss [t ha^{-1} yr^{-1}], that was predicted by the FAO project LADA (Land degradation assessment) (*Nachtergaele et al.*, 2011) with the Universal Soil Loss equation (USLE) (*Wischmeier & Smith*, 1978). c [ha yr t^{-1}] is an empirical coefficient that defines the relationship between the amount of TP in inorganic fertilizer, TP that is fixed to soil particles, and the amount of eroded soil (c was calibrated to c=3·10^{-6} ha yr t^{-1}).

WaterGAP3 calculates TP loadings from fertilizer application for the unsealed area in grid cells with cropland as dominant land use type. Hereby, it uses country-specific $F_{inorg,\mathrm{TP}}$ for 21 different crop types according to FAO data (*FAO*, 2003). These FAO data approximately represents the time period 1995 to 1999. However, between 1990 and 2010, the global application of phosphate in inorganic fertilizer increased by about 13% (*IFA*, 2014). Hence, IFA data were used to derive country-specific correction factors for the crop-specific fertilizer application relative to the period 1995-1999. These relative changes are averages for 5-years-intervals, i.e. 1990-1994, 1995-1999, 2000-2004, and 2005-2010.

3.1.6 Agriculture – livestock wastes

Phosphorus in livestock wastes is partly used as an organic fertilizer and is partly unused and remains in pastures. Either way it is partly absorbed by plants and what is not absorbed is largely washed off into surface waters. WorldQual calculates livestock TP production in each cell ($F_{org,\mathrm{TP}}$) as the product of an animal-specific TP excretion rate $ex_{i,\mathrm{TP}}$ and the number of animals ls_i:

$$F_{org,\mathrm{TP}}\,[\mathrm{t\ TP\ yr^{-1}}] = \sum_{i=1}^{12} ls_i ex_{i,TP} \tag{6}$$

The index i represents twelve animal types: dairy cattle, non-dairy cattle, pigs, sheep, goats, buffaloes, camels, horses, chicken, turkey, ducks, and geese. Animal type and density data were taken from the Water Use model of the WaterGAP3 modelling framework. The excretion rate for each specific animal type ex_i [t head^{-1} yr^{-1}] is computed using the approach of *Potter et al.* (2010):

$$ex_i[\text{t head}^{-1}\ \text{yr}^{-1}] = m_{TP,i} lc_i \tag{7}$$

in which $m_{TP,i}$ [t head^{-1} yr^{-1}] is the amount of TP in the manure of a particular type of animal (taken from *ASAE,* 2003, and therefore OECD data). The parameter lc_i [0-1] is animal-specific and accounts for regional differences in animal nutrition. These regional differences are taken into account by taking the ratio of regional and OECD equivalent livestock units. Values of lc are derived from *FAO* (2003) data.

Similar to $Ld_{if\text{TP}}$ [equation (5)], WorldQual assumes that the fraction $F_{org,\text{TP}}$ of wastes is washed out to the surface water system as a function of runoff:

$$Ld_{org,\text{TP}}[\text{t TP month}^{-1}] = F_{\text{org,TP}} \times \left(\frac{L_{max}}{1+(R_{act}/a)^{-b}} + Sl \times c \times \frac{R_{act}}{R_{mean}}\right) \times 1/12 \tag{8}$$

Again, the first and second terms within brackets represent leaching and erosion of total phosphorus, respectively.

3.1.7 Background loadings

WorldQual considers two background sources of TP. The first is atmospheric P deposition $F_{atm,\text{TP}}$ [t TP yr^{-1}], which was derived from the global distribution of deposition fluxes for TP in *Mahowald et al.* (2008). This deposition originates from both natural sources such as P contained in soils transported by wind and anthropogenic sources such as P contained in air pollution emissions. Based on these data, WorldQual estimates the fraction of atmospheric deposition that is washed out to the surface water as an empirical function of runoff:

$$Ld_{atm,\mathrm{TP}}[\mathrm{t\ TP\ month^{-1}}] = F_{atm,\mathrm{TP}} \times \left(\frac{L_{max}}{1 + (R_{act}/a)^{-b}} + Sl \times c \times \frac{R_{act}}{R_{mean}}\right) \times 1/12 \qquad (9)$$

The parameters L_{max}, a, b, and c are the same as for equations (5) and (8).

The second background source is chemical weathering of TP ($Ld_{cw,\mathrm{TP,annual}}$ [t TP yr^{-1}]). This involves the transport of P from soils and rock in the catchment via runoff. Data for this source were taken from *Hartmann et al.* (2014) and adjusted to monthly loadings using

$$Ld_{cw\mathrm{TP}}[\mathrm{t\ TP\ month^{-1}}] = Ld_{cw,\mathrm{TP,annual}} \frac{R_{act}}{R_{mean} \times 12} \qquad (10)$$

where R_{act} [mm yr^{-1}]) is the annual runoff, and R_{mean} [mm yr^{-1}] the mean annual runoff of the period 1980-2000. Runoff values are taken from the hydrological model of the WaterGAP3 modelling framework.

3.1.8 TP retention in the surface water system

Retention in surface water systems is defined by *Hejzlar et al.* (2009) as "the fraction of external N or P loading that is retained within the water bodies, either in absolute values or relative to the input". In WorldQual, retention of TP is calculated on the catchment scale based on the approach of *Behrendt & Opitz* (1999):

$$\frac{L_{rTP}}{L_{sumTP}} = \frac{1}{1 + a\,HL^{b}} \qquad (11)$$

Here, $L_{r\mathrm{TP}}$ is the TP loading at a catchment's outflow point, which in this study is equivalent to its lake inflow location. *HL* [m yr^{-1}] is the hydraulic load, defined as the ratio of annual runoff in the catchment [m^{3}yr^{-1}] to its water surface area [m^2] (*Hejzlar et al.*, 2009). Annual runoff is taken from the hydrology model of WaterGAP3 and surface area from *Alcamo et al.* (2003). The values for the empirical parameters a = 13.2 and b = -0.93 are taken from *Hejzlar et al.* (2009).

3.2 Reliability of the loading estimates

The reliability of the TP model can be tested by comparing calculations to measured TP loadings in 92 lake and river catchments (not equivalent to the 100 largest lakes), as well as the percentage share of each TP source (Table 2, Figure 3a). For studies with multi-year time series data [e.g. 1990-2000 for Lake Victoria as reported in *Scheren et al.* (2000)] we only use the long-term annual average of these data, rather than separate measurements from individual years. The references in Table 2 cover the available literature on TP loadings in the basins of large lakes and rivers according to the authors' knowledge.

An initial finding of this comparison with literature values is that the range of model and literature values is similar (Figure 3a). For individual basins, the nearest agreement is for the Danube basin (calculated vs measured, 30 vs. 31 kg km^{-2} yr^{-1}, respectively) and the farthest for Lake Taihu basin (calculated vs. measured, 384 vs. 65 kg km^{-2} yr^{-1}, respectively). The R^2 of model vs. measurements is 0.53, which is typical of other models in the literature [e.g. the NEWS-DIP model: R^2=0.55 (*Harrison et al.*, 2005)]. The mean absolute deviation of model vs measurements is 39%. On average, model estimates are 5 kg km^{-2} yr^{-1} higher than measurements.

As a further test of the model, we examine the consistency between WorldQual and other model estimates by comparing our TP source apportionment calculations with those of other models (Table 2). For this comparison we harmonize data from the different models into three categories: domestic + industry sector, agriculture sector, and background loadings (Figure 3b,c,d). We find that there is a good agreement between the average values of the model vs. literature values (Table 3), suggesting that WorldQual gives central estimates of P loadings with respect to the ensemble of available studies. There is a lower level of agreement when WorldQual is compared one-on-one with individual studies (Figures 3b, c, d). The model agrees with literature estimates moderately well for the domestic + industry sector, and less so for the agriculture sector and background loadings. The agreement between models for the domestic+industry sector suggests a similarity in methodology or input data for the different models.

Discrepancies between WorldQual and measurements or other model estimates may be due to various reasons. First, there observational and input data contain errors and uncertainties. Second, simplified model assumptions are a typical source of error in all models. In general, we believe the simplified representation of P processes in WorldQual is appropriate for global analysis, where the temporal and spatial resolution is coarse, and where the key objective is to obtain a consistent global overview of many different lakes. It is less appropriate for simulating local, specific conditions, e.g. the impact of particular aquaculture facilities on overall P loading to large lakes.

4 Results

To address the research questions presented at the beginning of this paper, we now analyze and compare the TP loading in the investigated 100 large lakes (Section 4.1), as well as their main sources (Section 4.2), and trends (Section 4.3). We present results for both TP loadings per catchment area and per lake area. The metric "loading per catchment area" illustrates the intensity of P produced in the catchment and is therefore useful for investigating strategies to reduce these loadings. The metric "loading per lake area" can be used as input to simplified assessments of eutrophication potential of lakes, e.g. along the lines of *Vollenweider*, (1976). Here we present mean annual loadings for two periods, 1990-1994 and 2005-2010 that are based on the sequentially modelled monthly loadings for the period 1990-2010. The five-year averaging period takes into account the temporal irregularity of inorganic fertilizer data (*IFA*, 2014). Since agriculture is a major source of TP loading, these data play a crucial role in model calculations.

Estimates of fertilizer use are based on country-specific fertilizer balances (production, consumption, production, export, and import) which do not necessarily reflect the actual use of fertilizer in a particular year. The problem is that countries frequently import fertilizer and store large quantities of it from year-to-year. In order to smooth out this effect we use five-year averaged data. Another reason for using five-year averaging periods is to smooth out the effect of year-to-year hydrologic variations.

4.1 Global distribution of TP loadings

For the world's 100 largest lakes, as defined in this paper, the median TP loading per catchment area (Figure 4a) was 5 kg km^{-2} yr^{-1} during the period 2005-2010 what is representing current conditions. The frequency distribution of TP loadings to these lakes has a positive skewness of 4.1, indicating a bias towards smaller loadings. Loadings are most frequently in the pollution class ranging from 0 to 1 kg km^{-2} yr^{-1}. Qinghai Lake in China has the largest loading (516 kg km^{-2} yr^{-1}), and the Canadian Lakes Dubawnt, Martre, and Nueltin in Canada have the lowest loadings (<0.01 kg km^{-2} yr^{-1}. Other large lakes in these countries also tend to be either very high or very low, respectively, in accordance with the density of population and economic activity, and level of environmental protection. Despite the large loadings to Chinese lakes, the Asian average is not as high as Latin America because of the numerous Siberian lakes with small loadings. The median of Latin America loadings is above the global average (70 kg km^{-2} yr^{-1}). In particular, Lake Titicaca stands out with loadings of about 115 kg km^{-2} yr^{-1}. African lakes (4 kg km^{-2} yr^{-1}) are below the median, and European lakes (median: 5 kg km^{-2} yr^{-1}) are close to the median of all lakes examined.

The distribution of TP loadings per lake area is similar to that of loadings per catchment area. Although spatial patterns are similar (Figure 4a and b), loadings per lake area tend to be two or more orders of magnitude larger because lake area is much smaller than lake catchment area. Anomalies are Europe and North America where loadings per lake area are only a factor of five and nine, respectively, larger than loadings per catchment area. This is because European and North American lakes have a smaller average ratio of catchment to lake surface. Because of this, TP loadings per lake area to African lakes are more than four times higher than to European lakes, although they have approximately the same loadings per catchment area (Table 4).

In the Northern Hemisphere, TP loadings to lakes tend to decrease with latitude, roughly in accordance with population density (Figure 5a). Nevertheless, lakes with high loadings can be observed at all latitudes. Below 30°N there is no apparent correlation between TP loadings and latitude or population density (Figure 5a).

Apart from a latitudinal variation, TP loadings also vary greatly between developing and developed countries (for definition of "developing" and "developed" see *UNEP*, 2017). Out of the 100 large lakes examined in this paper, 66 are located in developing countries, and these have an average TP loading of about 11.1 kg km^{-2} yr^{-1}. The remaining lakes are in developed countries and have a loading of about 0.7 kg km^{-2} yr^{-1}.

4.2 The sources of P loading

The sources of TP loadings to the large lakes investigated, in order of importance, are: inorganic fertilizer, background loadings (weathering and atmospheric deposition), domestic sewered wastewater, livestock wastes, and other sources [wastewater from manufacturing, domestic unsewered from scattered settlements (septic tanks, pit latrines, and others), and open defecation)]. However, the relative importance of these sources varies between lakes and continents as noted in following paragraphs.

Inorganic fertilizer accounts for about 47% of the total TP loadings to all large lakes surveyed, and more than 50% in 44 out of 100 large lakes. It accounts for 88% of TP loadings to large lakes in Asia (Figure 6c), ranging from <0.1% calculated for the remote Siberian lakes Taymyr and Vilyuyskoye to more than 99% for the lakes Hulun and Na-Mu (China) and Hovs Gol Lake (Mongolia). Inorganic fertilizer is also the most important source in Latin America (76%) with the exception of the Brokopondo Reservoir in Surinam, where only a small amount of inorganic fertilizer is applied. Inorganic fertilizer accounts for 45% of TP loadings in African lake catchments (Figure 6b), except for the Mai-Ndombe Lake (Congo) where it is estimated to be an unimportant source. In Europe, inorganic fertilizer accounts for 32% of TP loadings, ranging from 3% at Lake Vättern (Sweden) to 44% at Lake Peipsi (Estonia/Russia). In North America, inorganic fertilizer accounts for 28%, and it is a significant source in only about one-half of the lakes studied in this region. Many, but not all large lakes in Canada are in pristine condition, and a few (e.g. Lake of the Woods, 74%, and Williston Lake, 72%) receive TP loadings primarily from inorganic fertilizer.

As noted previously, *background loadings* are estimated here to be the second largest source of TP to large lakes, accounting for about 36%, globally. As expected they are most significant at remote locations. For example, background P accounts for the majority of TP loadings to most large Canadian lakes in thinly populated areas (Figure 6e), and is the sole source at several of these lakes (Lake Amadjuak, Lake Mistassini, Lake Martre, Lake Smallwood). Background P also accounts for most of the P input to some large lakes on other continents (e.g. (Lake Mai-Ndombe (Congo), 98%; Lake Taymyr (Russia), 98%; Vilyuyskoye Reservoir (Russia), 98%; Brokopondo Reservoir (Suriname), 95%).

The next largest source of TP loading is from *domestic sewered wastewater* which accounts for about 8% of the total. The largest contribution is in Europe, with 24% on average, ranging from 11% at Lake Vaenern, Sweden, to 39% at Lake Van, Turkey (Figure 6a). The latter is the largest percentage contribution to any of the large lakes investigated in this paper. Outside of Europe, domestic sewered wastewater is an important source of P to Lake Michigan in the United States (25%), Lake Chiquita in Argentina (35%), and Lakes Taihu (35%) and Kahanka (28%) in China.

Livestock wastes are the fourth largest source of TP loading, accounting for 6% of the total. This source accounts for 17% of the TP loading to large African lakes, and is among the top three sources to Lake Taihu (China), Lake Poyang (China), Zeyskoye (Russia), Boeng Tonle Chhma (Cambodia), Baharat ath Thartar (Iraq), and of almost all lakes in South America.

The remaining source categories – *wastewater from manufacturing, urban surface runoff, waste from scattered settlements (septic tanks, pit latrines, and others),* and *open defecation* –contribute in total only <3% to total TP loadings.

4.3 Trends in TP loading

The TP loadings to large lakes have not remained static over the past decades. We estimate that the 100 lakes investigated here have had a net increase of 15% between the periods 1990-1994 and 2005-2010, although trends for individual lakes and for continental totals vary greatly in direction and intensity (Figure 7).

TP loadings increased by 38% in North America, with significant differences between source sectors and lakes (Figure 8). For example, the total TP loading substantially increased in a small number of North American lake catchments due to inorganic fertilizer (e.g. Lake Winnipegosis +117%, Lake Manitoba +146%, Lake Cedar +96%, Southern Indian +121%), while background loadings increased and decreased the total loadings in pristine North American catchments (e.g.

Smallwood Reservoir, Canada, +46%; Lake Dubawnt, Canada, -51%). As previously noted, background loadings consist of atmospheric deposition and chemical weathering. According to our estimates, the variation in background loading in this cases were caused by differences in climatic conditions which led to more/less P release from chemical weathering and more/less wash-out of atmospheric deposition in 2005-2010 as compared to 1990-1994 (see equations 10 and 11). The change in climatic conditions could be due to long-term trend or year-to-year variability, or a combination of the two. While inorganic fertilizer and background loadings are increasing in some large lakes in North America, the majority of lakes on this continent show little or no change.

In Asia, the TP loading to large lakes has a net decrease of 18%. TP loadings have fallen substantially in large lakes in Central Asia and Siberia, because of a large decrease in loadings associated with inorganic fertilizer. Meanwhile, in other parts of Asia, loadings increase because of the upsurge in loadings associated with inorganic fertilizer and animal wastes.

European lake catchments show a decrease of about 35% in TP loading, mostly due to a strong decline in inorganic fertilizer loadings, but also because of reduced P from domestic sewered wastewater and livestock wastes.

African TP loadings have a net increase of 17%, although the trend in individual lakes is very different. For example, TP loadings increased by more than 100% to Lake Edward (Democratic Republic of Congo, Uganda) primarily because of increasing organic and inorganic fertilizer application. TP loadings decreased by about 85 % to Lake Rukwa (Tanzania) because of a massive drop in inorganic fertilizer application. An increase in loadings associated with animal wastes contributed to an overall increase in loading to many African lakes. Three lake catchments also experienced a negligible trend in loadings – Lake Bangweulu (Zambia), Lake Victoria (Tanzania, Uganda, Kenya), and Lake Volta (Ghana).

Out of all continents, TP loadings increased the greatest (+ 82%) in Latin America. The main cause was increased loadings associated with inorganic fertilizer.

Previously we noted that current TP loadings differ greatly between developing and developed countries. In contrast, we found that the relative number of lakes with up or downward trends is approximately the same in both, developed and developing countries: between 1990-1994 and 2005-2010 loadings increased in 50 out of 100 large lakes globally. This included *increases* in 30 out 66 in developing countries, and *decreases or no change* in 14 out of 44 in developed countries. Related to this, the model results show higher relative TP loading changes (between 1990-1994 and 2005-2010) in lower latitudes (Figure 5b).

5 Discussion

As noted in Section 4.1, we found that Total P loadings have a north to south increasing trend down to about 20ºN. This tendency is supported by results from individual lakes in the literature. For example, TP loadings to the northern Lake Ladoga, Russia (*Holopainen & Letanskaya*, 1999), Lake Onega, Russia (*Bilaletdin et al.*, 2011), Lake Michigan, USA (*Johengen et al.*, 1994), and Lake Winnipeg, Canada (*LWSB*, 2006) are relative low, compared to the southern Laguna de Bay, Philippines (*Zimmer & Bendoricchio*, 2001), Bara Bonita Reservoir, Brazil (*Salas & Martino*, 1991), and Lake Taihu, China (*Wang et al.*, 2014). However, there are exceptions, as in the case of the southern Lake Kivu (*Pasche et al.*, 2012) which has a low TP loading.

The north-south trend in TP loading to lakes is also consistent with *Seitzinger & Harrison* (2005) and *Bouwman et al.* (2009) who estimate that the P export of rivers (sum of dissolved inorganic, dissolved organic, and particulate P) tends to be lower in North America and the northern part of Asia than in Latin America and the southern part of Asia (part of Asia below 30°N), and Oceania.

Section 4.1 also notes the substantially higher loadings to lakes in developing countries as compared to developed countries. This is consistent with the well-known Environmental Kuznets Curve hypothesis that postulates an increase in environmental degradation along with income up to a certain point, after which environmental degradation begins to decrease (see *Dinda*, 2004).

As noted in Section 4.2, anthropogenic sources within a lake catchment dominate TP loadings to large lakes on the average. We estimate that anthropogenic sources in a lakes catchment account for about 73% of all P loadings (Table 3). Here we consider anthropogenic sources within a catchment. However, about 4.8 % in atmospheric phosphorus is from remote anthropogenic sources, on average (*Mahowald et al.*, 2008).

Both this paper and other estimates indicate that human sources are about three times higher than non-anthropogenic. This corresponds to an estimate of *Smil* (2000) that humans have nearly tripled the global phosphorus flows as compared to their natural levels. As an indicator of the degree of direct anthropogenic influence on the P loading to each large lake we compute the following:

$$I_{TP}[0,1] = L_h / L_{h+b} \qquad (12)$$

with L_h (mean annual loading from direct anthropogenic sources) and L_{h+b} (the sum of mean annual TP from anthropogenic sources in the catchment plus background). As a degree of anthropogenic influence, we set $I_{TP} < 0.5$ as background dominated, and $I_{TP} > 0.5$ as human dominated TP sources (if both are 0.5, there is no dominant source). Applying this impact indicator to the results, it is indicating that about 74 of the 100 studied lake catchments exhibit human dominated TP sources (Figure 9).

The trends in TP loading were ultimately driven by social and political change. For example, the literature provides some answers as to why fertilizer application (which had such a strong influence on the trend of loading) increased in the south and decreased in the north. In the 1990s Russian agriculture was restructured and agricultural production collapsed during the transition from the collective farm system to a market economy (*Brooks & Gardener*, 2004). During this time total phosphate consumption in countries of the in the former Soviet Union decreased rapidly from 4.3 million tons in 1990 to 0.26 million tons in 1994 (*IFA*, 2014). This is a likely explanation for the clear decrease of TP loading to large lakes in Central Asia and Siberia. Meanwhile, TP loadings to lakes in Central and Western Europe decreased due to investments in water pollution control. The average of loadings to southern lakes (south of 40°N) increased due to several reasons. One reason was strong economic growth in China, which is the world's largest consumer of synthetic fertilizer (*Gao et al.*, 2006). Fertilizer application in China, in terms of tons of P_2O_5 , increased from 5.8 to 12.1 between 1990 and 2010 (*IFA*, 2014). An increase in the eutrophication of lakes was also observed during this period (*Zhou et al.*, 2014). Meanwhile, in Latin America, P_2O_5 use in agriculture increased from 2.3 to 5.1 million tons between 1990 and 2010 (*IFA*, 2014).

6 Conclusions

Total phosphorus (TP) loading is known to play a key role in the eutrophication of lakes. In this paper we have used the WorldQual model to investigate the TP loading to 100 of the largest lakes in the world. An advantage of using a single model is that it provides consistent and comparable estimates for all 100 lakes. A disadvantage is that it does not provide the robustness of multi-model estimates. Also the model used is better suited for global-scale calculations and less appropriate for catchment-specific studies.

From our analysis, we find, first, that the TP loadings to large lakes in developing countries are on the average much larger than to lakes in developed countries (11.1 vs. 0.7 kg km^{-2} yr^{-1}). Second, we estimate that the main source of TP loadings is inorganic fertilizer. The second most important source is the background loading from (1) atmospheric deposition of P from anthropogenic and non-anthropogenic sources, and (2) P released through rock and soil weathering in the catchment. Third, TP loadings have increased between 1990-1994 and 2005-2010 in 50 of the 100 large lakes investigated. The main cause of increasing or decreasing TP Loadings was the trend in inorganic fertilizer use in lake catchments.

In sum our findings indicate great differences in the TP loading and, hence, the eutrophication potential between the world's largest lakes. In particular, the risk of P-stimulated eutrophication is much larger in developing countries than developed countries, and this justifies greater action especially in developing countries to reduce these loadings and avoid losing the valuable ecosystem services provided to society by these large lakes.

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Figures

Figure 1: Assignment of countries to continents used in this paper. Lighter orange = Central and North America; darker orange = South America; lighter blue = Europe; darker blue = Africa; darker green = Asia; darker green = Oceania. The points indicate the positions of the investigated lakes.

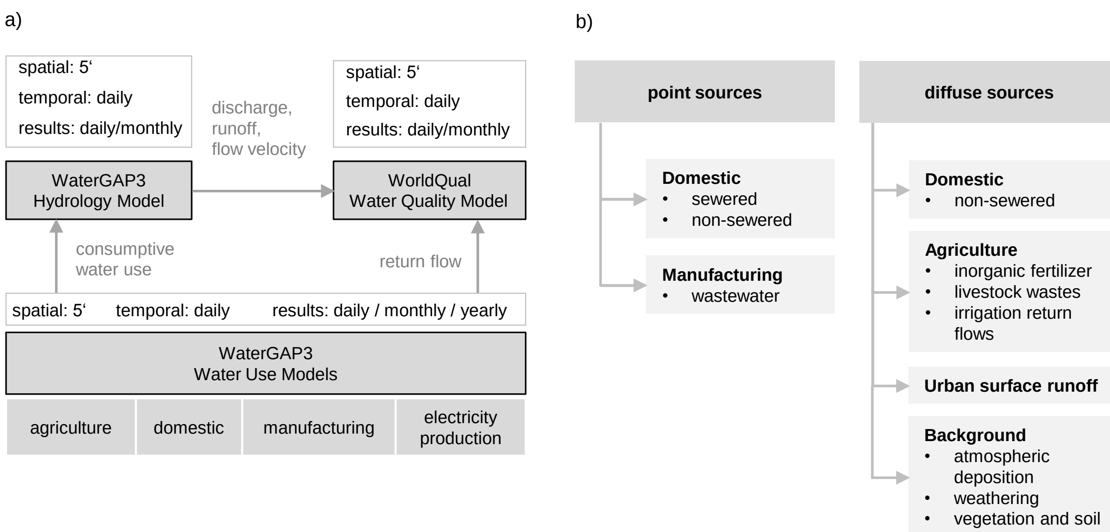

Figure 2: a) Overview of the WaterGAP3 modelling framework (*Verzano*, 2009; modified), and b) water pollutant loading sectors in WorldQual categorized as either point sources or diffuse sources.

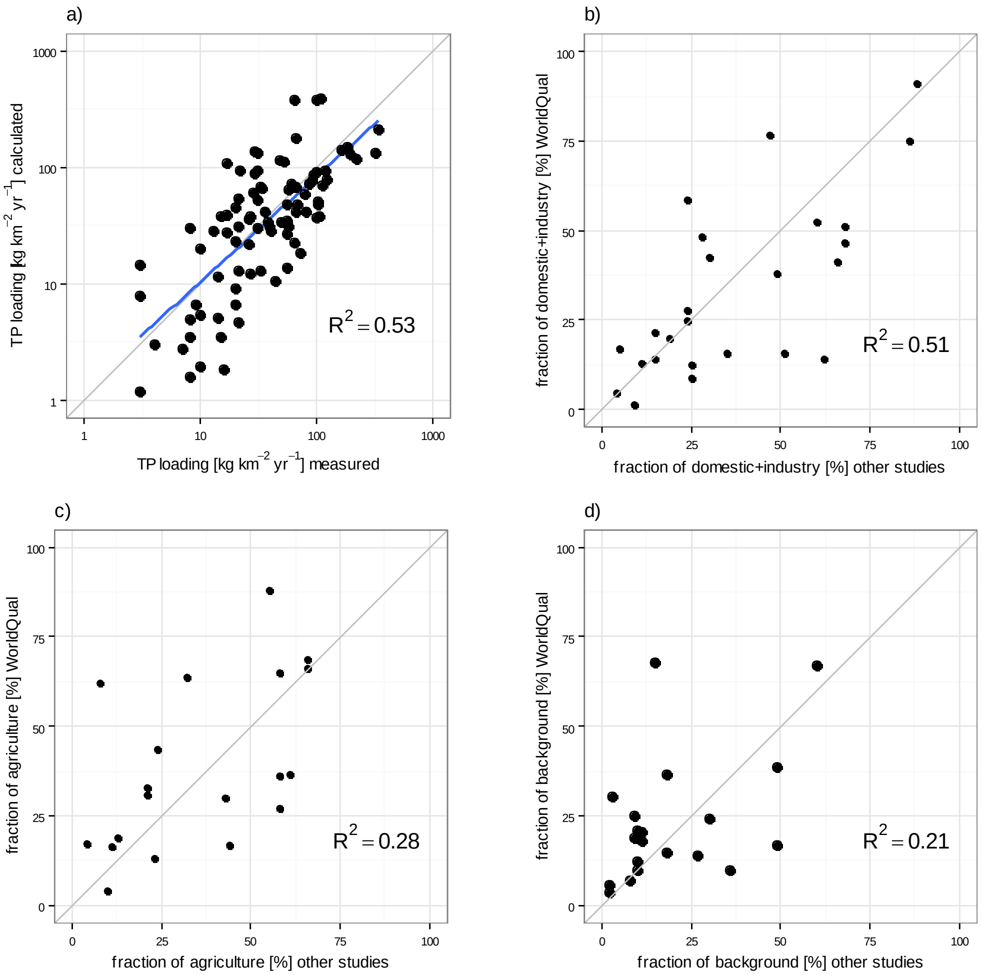

Figure 3: Testing of total WorldQual TP calculations. The loadings in (a) are given per unit catchment area. Diagrams b) to d) show modeled sectoral contributions vs. data of other source apportionment studies (no evaluation).

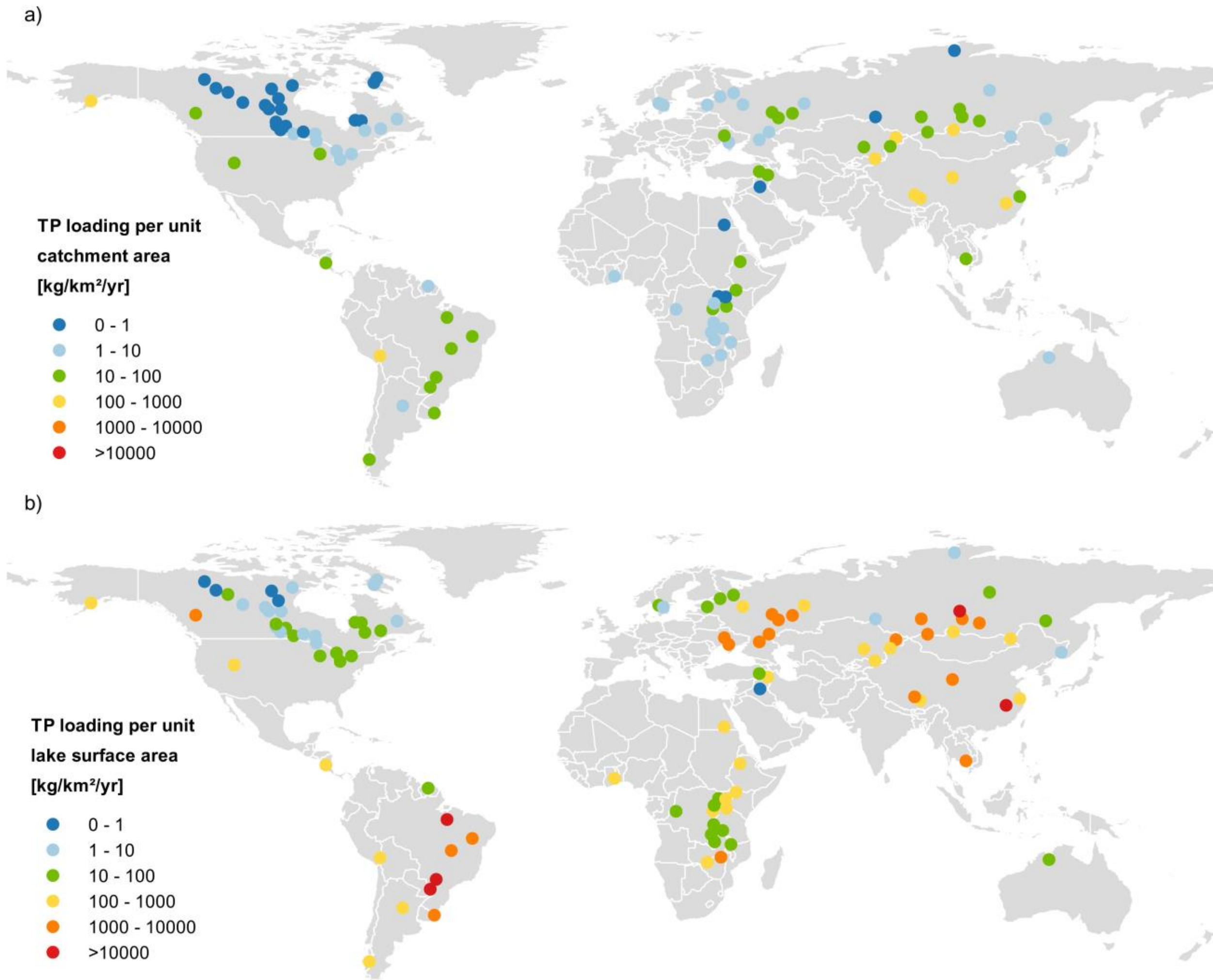

Figure 4: Total phosphorus loadings to the 100 largest lakes in the world with respect to catchment size, for the period 2005-2010: a) loadings per unit catchment area, and b) loadings per unit lake surface area.

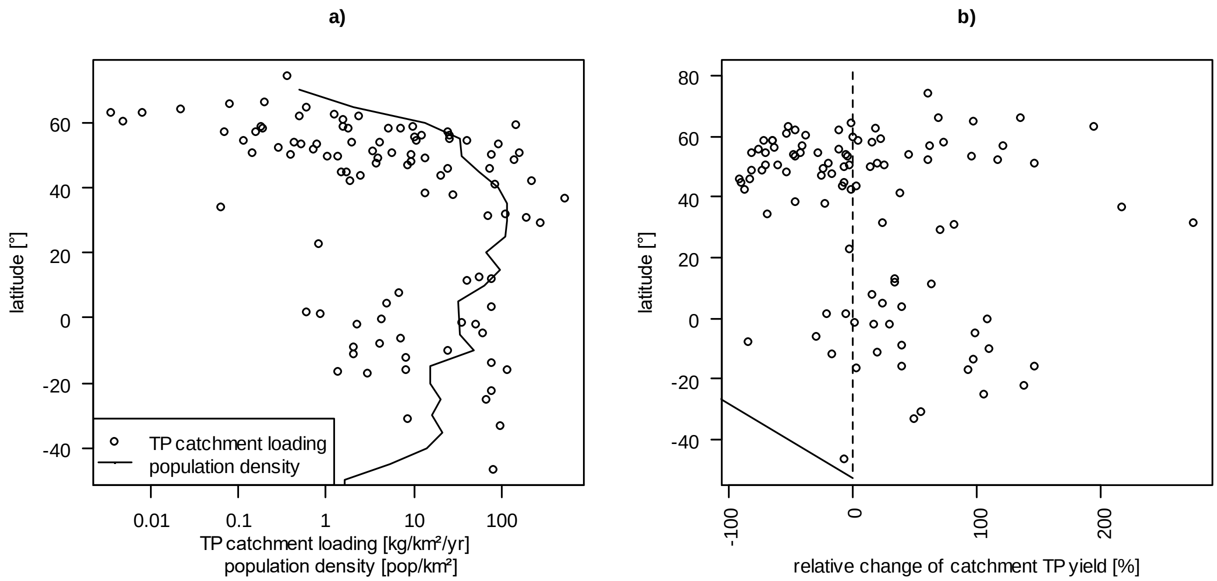

Figure 5: Latitudinal dependence of (a) TP catchment loading (WorldQual calculations) and population density (*Kummu & Varis*, 2011), and (b) the change of calculated loading between 1990-1994 and 2005-2010.

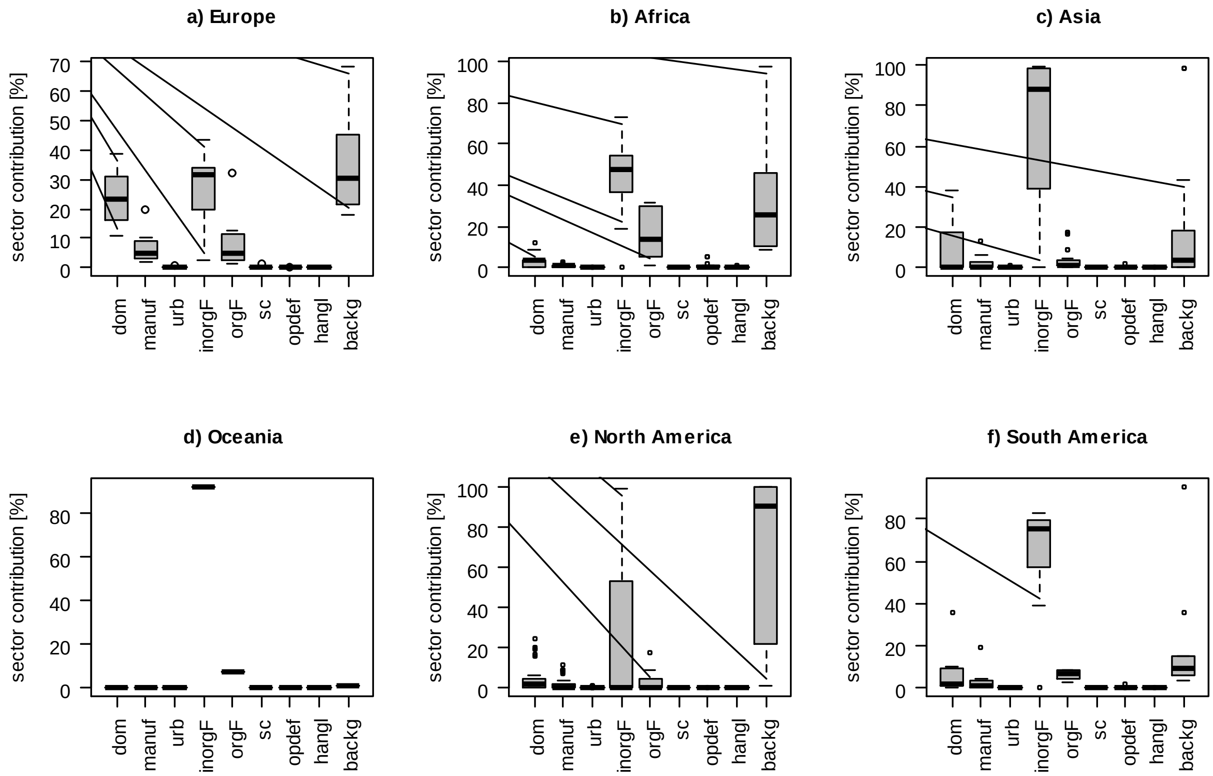

Figure 6: Sectoral contribution to total phosphorus loadings in the catchments of the hundred largest lakes, for the period 2005-2010, and for each continent. dom = domestic sewered wastewater, manuf = wastewater from manufacturing, urb = urban surface runoff, inorgF = inorganic fertilizer, orgF = livestock wastes, sc= domestic unsewered wastes from scattered settlements (septic tank, pit latrines and others), opdef = open defecation, hangl = hanging latrines, backg = background (weathering and atmospheric deposition).

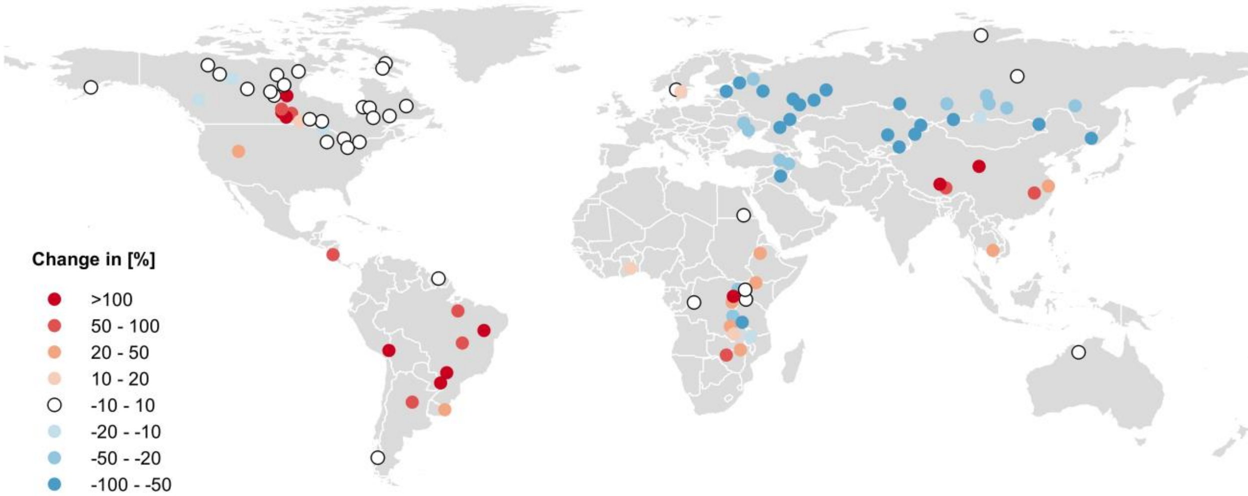

Figure 7: Changes of mean annual TP loadings to the world's 100 largest lakes between the periods 1990-1994 and 2005-2010. To filter out variations of very low loadings to remote lakes, all changes were set to 0 if the fraction of background loadings is larger than 90%.

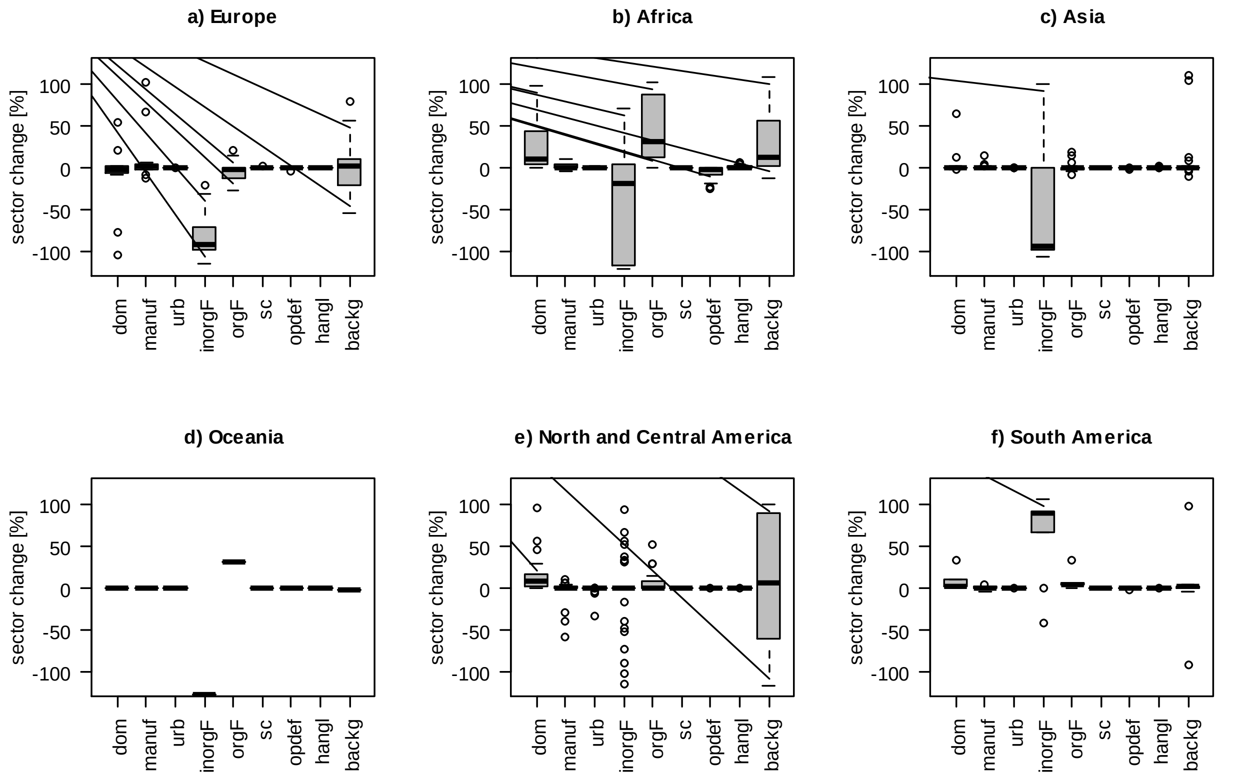

Figure 8: Changes of mean annual loadings from each sector relative to the sum of changes in all sectors between the periods 1990-1994 and 2005-2010 for each continent. dom = domestic sewered wastewater, manuf = wastewater from manufacturing, urb = urban surface runoff, inorgF = inorganic fertilizer, orgF = livestock wastes, sc= domestic unsewered wastes from scattered settlements (septic tank, pit latrines and others), opdef = open defecation, hangl = hanging latrines, backg = background (weathering and atmospheric deposition).

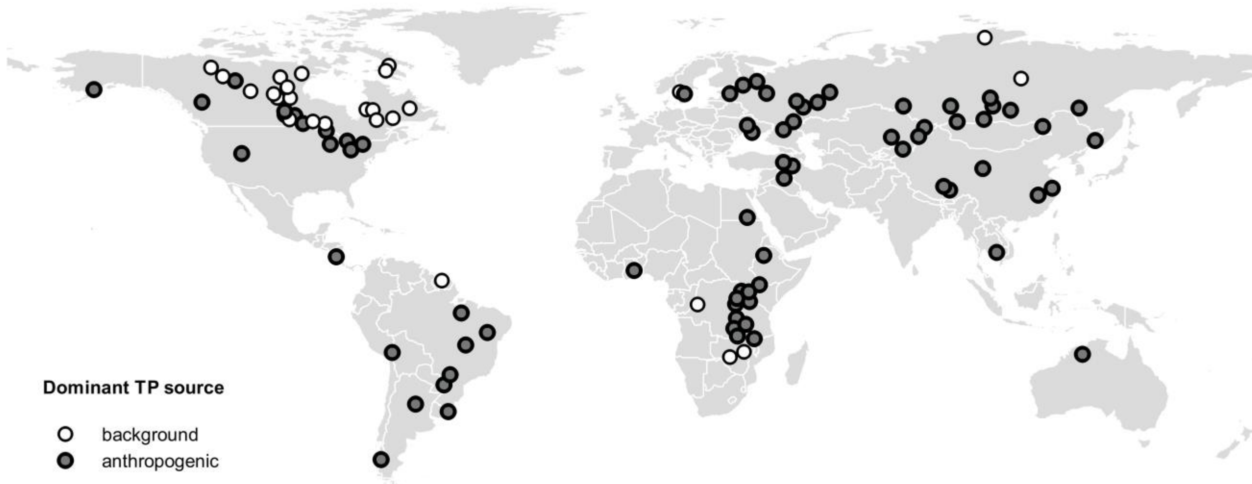

Figure 9: Dominant TP source in a lake catchment. The dominant source accounts for more than 50% of the direct TP loadings.

Tables

Table 1: Compilation of global data sources used in TP loading calculations

Data	Application	Data source or related publication	Original spatial resolution
Protein consumption data	domestic sewered and non-sewered	*FAO* (2014)	countries
Population	domestic sewered and non-sewered	*Klein Goldewijk* (2005), *Klein Goldewijk et al.*, (2010)	5 arc minutes
Treatment level	domestic sewered, manufacturing wastewater, urban surface runoff	*Williams et al.* (2012)	countries
Sanitation practice (scattered settlements)	domestic non-sewered	*WHO/UNICEF* (2013)	countries
Fertilizer application by crop	inorganic fertilizer	*FAO* (2003)	countries
Total fertilizer application	inorganic fertilizer	*IFA* (2014)	countries
Soil loss	all diffuse sources	*Nachtergaele et al.* (2011)	5 arc minutes
Animal type and density	livestock wastes	*FAOSTAT* (2014)	countries
Phosphorus in manure	livestock wastes	*ASAE* (2003)	countries
Livestock units	livestock wastes	*FAO* (2003)	FAO regions
Chemical weathering	background loads	*Hartmann et al.* (2014)	0.008°
Atmospheric deposition	background loads	*Mahowald et al.* (2008)	0.5°
Input from the WaterGAP3 hydrology model			
Urban surface runoff	urban surface runoff	WaterGAP3 calculations published in *Schellekens et al.* (2017)	5 arc minutes
Built up fraction	urban surface runoff retention calculation	WaterGAP3 standard input (*Alcamo et al.*, 2003)	5 arc minutes
Water surface area	retention calculation	WaterGAP3 standard input (*Alcamo et al.*, 2003)	5 arc minutes
Surface runoff	all diffuse sources	WaterGAP3 calculations published in *Schellekens et al.* (2017)	5 arc minutes

Table 2: List of references used for model testing

Reference	River and lake basins	Number of data points
TP loading data		
Meybeck & Ragu (1995)	Large river discharges into the ocean.	15
OECD (2015)	Kymijoki River	1
Pedrozo & Bonetto (1989)	Parana River	1
Skogen & Soiland (2001)	Rivers into the north sea	3
Lurry & Dunn (1997)	Mississippi River Basin	1
Stalnacke et al. (1999)	Rivers into the Baltic Sea	3
de Wit (2000)	Elbe and Rhine basins	3
LfW (2002)	Mosel River	1
Teodoru & Wehrli (2005)	Danube at inflow to Black Sea and Iron Gate Reservoir	3
Saunders & Lewis (1988)	Apure River	1
Chinese Academy of Science (2011)	Yangtze River	1
Skoulikidis et al. (1998)	Greece Rivers	6
Ludwig et al. (2009)	Rivers into the Mediterranean and Black Sea	19
Holopainen & Letanskaya (1999)	Lake Ladoga	1
Bilaletdin et al. (2011)	Lake Onega	1
ILEC (2015)	Lakes of the world	11
Scheren et al. (2000)	Lake Victoria	1
Zimmer & Bendoricchio (2001)	Laguna de Bay	1
Johengen et al. (1994)	Lake Michigan and Ontario	2
Dolan & McGunagle (2005)	Lake Erie	1
LWSB (2006)	Lake Winnipeg	1
IGKB (2000)	Lake Constance	1
Pasche et al. (2012)	Lake Kivu and Malawi	2
Matzinger et al. (2007)	Lake Ohrid	1
Wang et al. (2014)	Lake Taihu	1
de Anda et al. (2001)	Lake Chapala	1
Salas & Martino (1991)	South American Lakes	3
European Environment Agency (2005)	Large European Lakes	3
BUWAL (1994)	Swiss Lakes	3
	Total	**92**
Data on sectoral contribution*		
European Environment Agency (2005)	European Rivers and Lakes	10
White & Hammond (2011)	Thames	1
LfW (2002)	Mosel River	1
ILEC (2015)	Lakes of the world	9
Scheren et al. (2000)	Lake Victoria	1
LWSB (2006)	Lake Winnipeg	1
IGKB (2000)	Lake Constance	1
	Total	**24**

* The references do not provide the fraction of contribution of each sector for all lakes and rivers. Thus, each subplot b-d in Figure 3 does not contain 24 data points in total.

Table 3: Comparison of estimates (WaterGAP3 and other studies) for average percentage share of TP sources.

Source	Average percentage share calculated in this paper [%]	Average percentage share of other studies [%]
Domestic + industry	33.2	37.5
Agriculture	38.7	35.6
Background	23.0	19.4

Table 4: Continental median, maximum, and minimum of mean annual TP yields and loadings for the period 2005-2010.

	Global	Europe	Africa	Asia	Oceania	North and Central Amerika	South America
TP loadings of the lake catchment							
median [kg km^{-2} yr^{-1}]	5	5	4	25	(1) [a]	1	70
max [kg km^{-2} yr^{-1}]	516	28	75	516	-	143	115
min [kg km^{-2} yr^{-1}]	0	0	1	0	-	0	5
TP loading relative to the lake surface area							
median [kg km^{-2} yr^{-1}]	94	29	136	1037	(36) [a]	9	1231
max [kg km^{-2} yr^{-1}]	23180 [b]	2628	1914	20640 [b]	-	1089	23180 [b]
min [kg km^{-2} yr^{-1}]	0	1	22	2	-	0	36

[a] This is the average annual TP yield in the Argyle Reservoir catchment (2005-2010) which is the only lake investigated in Oceania. Therefore no statistics presented.
[b] See the evaluation of these high estimates in the Discussion section.